# Novel nonreciprocal acoustic effects in antiferromagnets


Roser Valentí, Claudius Gros

*Institut für Physik, Universität Dortmund, 44221 Dortmund, Germany.*

V.N. Muthukumar

*Department of Physics, Brookhaven National Laboratory, Upton, New York 11973-5000*



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## Abstract


The possible occurrence of nonreciprocal acoustic effects in antiferromagnets in the absence of an external magnetic field is investigated using both (i) a microscopic formulation of the magnetoelastic interaction between spins and phonons and (ii) symmetry arguments. We predict for certain antiferromagnets the existence of two new nonreciprocal (non-time invariant) effects: A boundary-condition induced nonreciprocal effect and the occurrence of transversal phonon modes propagating in opposite directions having different velocities. Estimates are given and possible materials for these effects to be observed are suggested.



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*Introduction* It is well known in optics that the coupling of light to the order parameter in a ferromagnet mediates nonreciprocal effects such as the Faraday effect [1]. The Faraday effect in ferromagnets appears as a rotation of the plane of polarization of light which is incident along the axis of magnetization. An analogous effect in acoustics (the so-called *acoustic* Faraday effect) was first predicted by Kittel [2]. Subsequently, it was verified experimentally [3] that in a ferromagnet, circularly polarized phonons of different sense have

different velocities when the direction of propagation is parallel to the axis of magnetization. Such an effect can be understood in terms of the magnetoelastic coupling between spins and phonons. Boiteux *et al.* [4] performed in 1971 a detailed study of the magnetoacoustic effects in antiferromagnetic $Cr_2O_3$. They found that the crystal symmetry of $Cr_2O_3$ allows an acoustic Faraday effect only in the presence of an external magnetic field.

The nonreciprocal effects described above rely on the existence of a well defined magnetic moment induced either by the internal moments in the case of ferromagnets, or by the application of an external magnetic field in the case of $Cr_2O_3$ [4]. It is not apparent that such effects can occur in ordered antiferromagnets in the absence of an external magnetic field where the total magnetic moment is zero. In the case of optics, symmetry arguments can be used to deduce the presence of nonreciprocal bulk effects [5] as well as a range of characteristic surface effects [6] in antiferromagnets. Indeed, nonreciprocal effects in antiferromagnets were demonstrated recently using the technique of Second Harmonic Generation [7]. A microscopic explanation of these experiments which relies on the symmetry of the crystal and the evaluation of the interactions at the atomic level has also been proposed [8,9].

In this Letter we examine if it is possible to observe nonreciprocal phonons in antiferromagnets in the absence of an external magnetic field. We find that the answer is affirmative but these effects are very different from those observed in optics. Our calculations show that there is a new boundary-condition induced elliptical-polarization effect in antiferromagnets. In addition we predict, for certain antiferromagnets, the occurrence of transversal phonon modes propagating in opposite directions having different velocities, an effect which cannot occur in ferromagnets.

*Ferromagnets* Kittel [2] was the first to predict the magnetoacoustic Faraday effect in ferromagnets within a semiclassical continuum formulation. For the sake of completeness, we present a brief derivation of Kittel's result within a linear chain model for a ferromagnet. We take the $\hat{z}$-axis to be the direction of the chain of ions corresponding to the main crystallographic axis, which we assume to have $C_4$ or higher rotational symmetry. The magnetization is parallel to the $\hat{z}$-axis.

We define $u_{n,\gamma}$ and $p_{n,\gamma}$ ($n$ denotes the site index and $\gamma = x, y$) to be the canonical displacement and momentum operators obeying the commutation relations $[u_{n,\gamma}, p_{n',\gamma'}] = i\hbar\delta_{n,n'}\delta_{\gamma,\gamma'}$. The Hamiltonian $H = T + V_{el} + H_{Heis} + H_{sp}$ contains the unperturbed phonon part

$$T = \frac{1}{2m}\sum_{n,\gamma} p_{n,\gamma}^2, \qquad V_{el} = \frac{K}{2}\sum_{n,\gamma}(u_{n+1,\gamma} - u_{n,\gamma})^2 \tag{1}$$

where $K$ is the elastic constant, and $m$ the mass of the ion. $H_{Heis}$ corresponds to the Heisenberg Hamiltonian and describes the unperturbed spin-spin interaction,

$$H_{Heis} = J\sum_n \vec{S}_n \cdot \vec{S}_{n+1} \tag{2}$$

$J < 0$ is the ferromagnetic exchange constant and $\vec{S}_n$ corresponds to the spin operator on site $n$, with $[S_n^\alpha, S_m^\beta] = i\hbar\delta_{n,m}\epsilon_{\alpha\beta\gamma}S_n^\gamma$ ($\alpha, \beta, \gamma = x, y, z$). $H_{sp}$ describes the magnetoelastic coupling between phonons and spins:

$$H_{sp} = g\sum_{n,\gamma}(u_{n+1,\gamma} - u_{n,\gamma})\left(S_{n+1}^\gamma S_n^z + S_{n+1}^z S_n^\gamma\right). \tag{3}$$

$g$ is the magnetoelastic coupling constant.

Eq. (3) is rotational invariant around the $\hat{z}$-axis. A 90-degree rotation around the $\hat{z}$-axis would correspond to ($u_{n,x} \to -u_{n,y}$, $u_{n,y} \to u_{n,x}$, $S_n^x \to -S_n^y$, $S_n^y \to S_n^x$). Eq. (3) is the lattice version of the usual coupling between components of the strain tensor and the magnetization [2]. This interaction term can also be understood as a higher order expansion of the exchange term and has its origin in the spin-orbit coupling [10]. In order to obtain the time dependence of the variables $p_{n,\gamma}$, $u_{n,\gamma}$ and $S_n^\gamma$ we consider the Heisenberg equation of motion $i\hbar\frac{d}{dt}A = [A, H]$. For simplicity, we work with circular-polarized coordinates $u_n^\pm = u_{n,x} \pm iu_{n,y}$, $S_n^\pm = S_n^x \pm iS_n^y$ and we assume a plane wave ansatz $u_n^\pm(t) = \exp[i(\omega t - nk)]u^\pm$, $S_n^\pm(t) = \exp[i(\omega t - nk)]S^\pm$ (the unit cell constant is taken to be one) in order to solve the equations of motion. We set the magnetization to be along the $\hat{z}$-axis ($\langle S_n^x\rangle = 0$, $\langle S_n^y\rangle = 0$, $\langle S_n^z\rangle = \langle S^z\rangle$) and we obtain after eliminating $\dot{p}_{n,\gamma}$:

$$\omega S^\pm = \pm g\langle S^z\rangle^2(2i\sin k)u^\pm \pm J\langle S^z\rangle 2(1 - \cos k)S^\pm \tag{4}$$

$$(-m\omega^2 + 2K(1 - \cos k))u^\pm = g\langle S^z\rangle(-2i\sin k)S^\pm\ . \tag{5}$$

Combining Eq. (4) and (5) we get:

$$\left\{[-m\omega^2 + 2K(1-\cos k)][\omega \mp J\langle S^z\rangle 2(1-\cos k)] \mp 4g^2\langle S^z\rangle^3(\sin k)^2\right\} u^{\pm} = 0 \tag{6}$$

which describes the dispersion relation of the phonon system interacting with the spin system. Note that for $u^+$ and $u^-$ the dispersion relation is different implying that for a given frequency $\omega$, right (+) and left (-) circular polarized phonons have different wave vectors $k^+$ and $k^-$ respectively.

An incoming linear polarized wave with frequency $\omega$ is decomposed into a linear superposition of right and left circular polarized waves with the same frequency $\omega$ but different wave vectors $k^+/k^-$ via $\exp[i(\omega t - k^+ n)]\,\mathbf{u}^+ + \exp[i(\omega t - k^- n)]\,\mathbf{u}^-$. The physical amplitudes, determined by the real part of $u$ are given by

$$\begin{aligned} u_{n,x} &= u\cos(\omega t - n(k^+ + k^-)/2)\cos(n(k^- - k^+)/2) \\ u_{n,y} &= u\cos(\omega t - n(k^+ + k^-)/2)\sin(n(k^- - k^+)/2). \end{aligned}$$

The plane of polarization rotates around the $\hat{z}$-axis [2] and it is easy to obtain from the solution of Eq. (6) the rotatory power $(k^+ - k^-)/(k^+ + k^-) \approx g^2\langle S^z\rangle^3/(K\omega)$. The nonreciprocal behavior is a direct consequence of the coupling between phonons and spins in a ferromagnet. For a typical ferrite the rotatory power should be of the order of 1/4 for sound waves in the microwave regime ($w \approx 10^9 s^-1$), as was corroborated experimentally [3].

*Antiferromagnets* As a model for an antiferromagnetic material, we consider a linear chain with two magnetic ions ($A/B$) per unit-cell, with $\vec{u}_{A,n}$ and $\vec{u}_{B,n}$ being their respective displacement vectors. A straightforward generalization of (1), (2) and (3) leads to

$$\begin{aligned} T &= \frac{1}{2m}\sum_{n,\gamma}\left(p_{A,n,\gamma}^2 + p_{B,n,\gamma}^2\right) \\ V_{el} &= \frac{K_1}{2}\sum_{n,\gamma}(u_{B,n,\gamma} - u_{A,n,\gamma})^2 + \frac{K_2}{2}\sum_{n,\gamma}(u_{A,n+1,\gamma} - u_{B,n,\gamma})^2 \ . \end{aligned} \tag{7}$$

for the phonon part. $K_1$ and $K_2$ are, respectively, the intra- and intercell elastic constants.

$$H_{Heis} = \sum_n \left[J_1\vec{S}_{A,n}\cdot\vec{S}_{B,n} + J_2\vec{S}_{B,n}\cdot\vec{S}_{A,n+1}\right] \ , \tag{8}$$

corresponds to the Heisenberg term with $J_1 > 0$ and $J_2 > 0$ (antiferromagnetic). And the magnetoelastic coupling is given by

$$H_{sp}^{(g)} = g_1 \sum_{n,\gamma} (u_{B,n,\gamma} - u_{A,n,\gamma})(S_{B,n}^{\gamma} S_{A,n}^{z} + S_{B,n}^{z} S_{A,n}^{\gamma})$$
$$+ g_2 \sum_{n,\gamma} (u_{A,n+1,\gamma} - u_{B,n,\gamma})(S_{A,n+1}^{\gamma} S_{B,n}^{z} + S_{A,n+1}^{z} S_{B,n}^{\gamma}) \tag{9}$$

where $g_1$ and $g_2$ are, respectively, intra- and intercell magnetoelastic coupling constants. In order to obtain the dispersion relation of the phonons interacting with the spin system, we set the staggered magnetization to be parallel to the $\hat{z}$ axis ($\langle S_{A,n}^z \rangle \to \langle S_A^z \rangle$ and $\langle S_{B,n}^z \rangle \to -\langle S_A^z \rangle$) and solve the equations of motion for $\vec{u}_{A/B,n}$, $\vec{p}_{A/B,n}$ and $\vec{S}_{A/B,n}$.

We consider the plane-wave ansatz $\mathbf{u}_n = \exp[i(\omega t - kn)]\,\mathbf{u}$ and define the vector $\mathbf{u}_n = (u_{A,n,x}, u_{B,n,x}; u_{A,n,y}, u_{B,n,y})$. Eliminating $\dot{p}_{A/B,n,\gamma}$ and $\dot{S}_{A/B,n,\gamma}$ we obtain $M\mathbf{u} = 0$, where the $4 \times 4$ hermitian matrix $M$ is given by:

$$\begin{pmatrix} L(\omega) + 2Js^4 R_k & -T_k(\omega) - Js^4 P_k & 2is^3 Q_k \omega & 0 \\ -T_{-k}(\omega) - Js^4 P_{-k} & L(\omega) + 2Js^4 R_k & 0 & -2is^3 Q_k \omega \\ -2is^3 Q_k \omega & 0 & L(\omega) + 2Js^4 R_k & -T_k(\omega) - Js^4 P_k \\ 0 & 2is^3 Q_k \omega & -T_{-k}(\omega) - Js^4 P_{-k} & L(\omega) + 2Js^4 R_k \end{pmatrix}$$

$s$ stands for $\langle S_A^z \rangle$, $\omega_0^2 = 4J^2 s^2 \sin^2(k/2)$. We consider for simplicity $J_1 = J_2 = J$ and the following definitions are used:

$$Q_k = g_1 g_2 (1 - \cos k)$$
$$R_k = (g_1^2 + g_2^2 - 4g_1 g_2)(1 + \cos k) + 2(g_1^2 + g_2^2)$$
$$P_k = (1 + \exp[-ik])(g_1 - g_2 \exp[ik])^2 +$$
$$(1 + \exp[ik])(g_1 - g_2)^2 + 4(g_1 - g_2)(g_1 - g_2 \exp[ik])$$
$$L(\omega) = (-m\omega^2 + K_1 + K_2)(\omega^2 - \omega_0^2)$$
$$T_k(\omega) = (K_1 + K_2 \exp[ik])(\omega^2 - \omega_0^2) \tag{10}$$

The eigenvectors of $M$ are given by:

$$\mathbf{u}^{+} = (u_A, u_B; iu_A, iu_B) \tag{11}$$

$$\mathbf{u}^{-} = (u_B^*, u_A^*; -iu_B^*, -iu_A^*) \tag{12}$$

and the amplitudes $u_A$ and $u_B$ are determined by

$$\{L(\omega) - 2\omega s^3 Q_k + 2Js^4 R_k\}u_A = \{T_k(\omega) + Js^4 P_k\}u_B \tag{13}$$

The dispersion relation $\omega = \omega(k)$ is given by the solution of $detM = 0$. $\mathbf{u}^+$ and $\mathbf{u}^-$ have the same dispersion what translates to the fact that right and left circular polarized phonons are degenerate and one might think, that there are no nonreciprocal effects in antiferromagnets. This degeneracy is broken when an external magnetic field is applied (see [4]). We shall show here that, in fact, even in the absence of an external magnetic field, nonreciprocal effects might be obtained by considering an appropriate choice of boundary conditions.

Let us consider a system that is driven by a linear polarized amplitude in the $\hat{x}$-direction at site $n = 0$; *i.e.*, we apply the external driving force

$$u_{A,0,x} = \exp[i\omega t]\, u^{(ext.)}, \qquad u_{A,0,y} \equiv 0. \tag{14}$$

The appropriate superposition of right and left circular polarized acoustic waves $\mathbf{u}^+/\mathbf{u}^-$ that satisfy (14) is

$$\mathbf{u} \equiv u_B^*\,\mathbf{u}^+ + u_A\,\mathbf{u}^- = (2u_B^* u_A, |u_B|^2 + |u_A|^2; 0, i(|u_B|^2 - |u_A|^2)),$$

with $2u_B^* u_A = u^{(ext.)}$. The physical amplitudes, given by the real part of $\exp[i(\omega t - kn)]\mathbf{u}$, are therefore linearly polarized for the $A$-site,

$$u_{A,n,x} = \cos(\omega t - kn)\, u^{(ext.)}, \qquad u_{A,n,y} = 0,$$

and elliptically polarized for the $B$-sites:

$$u_{B,n,x} = \cos(\omega t - kn)\,(|u_A|^2 + |u_B|^2),$$

$$u_{B,n,y} = \sin(\omega t - kn)\,(|u_A|^2 - |u_B|^2).$$

The degree of the *homogeneous* elliptical polarization $(|u_A|^2 - |u_B|^2)/(|u_A|^2 + |u_B|^2) \sim \langle S_A^z\rangle^3 g_1 g_2(1-cosk)/(m\omega^3)$ is proportional to the spin-phonon couplings $g_1$, $g_2$ and to $\langle S_A^z\rangle$, which makes it nonreciprocal. Therefore, by choosing appropriate boundary conditions it is possible to observe this nonreciprocal effect in an antiferromagnet. Note, in this case, the nonreciprocity relates to the amplitudes of the sound wave and not to the velocity as it happens in the ordinary acoustic Faraday effect in ferromagnets.

Since the magnetoelastic coupling between phonons and spins is determined by general symmetry arguments (see also [2] and [4]) that are not constrained by the dimensionality, we can extend our analysis to two and three dimensional materials. Then, it follows that the boundary-condition induced nonreciprocal effect should occur in materials that consist of antiferromagnetically stacked chains or planes with each chain/plane having a non vanishing moment along the stacking direction. Typical examples are compounds belonging to the family of $ABX_3$-type hexagonal antiferromagnets, like $CsCoCl_3$ and $CsNiBr_3$ [11]. The magnitude of the effect should be comparable to the nonreciprocal effect observed in ferromagnets [2,3] since both effects originate from the same microscopic coupling mechanism.

*Novel effects* In the case illustrated above, we considered a system of ions aligned along the $\hat{z}$-axis with cubic symmetry in the paramagnetic phase. Let us now assume a crystal with lower symmetry than cubic which also orders antiferromagnetically. We add to our Hamiltonian (9) the following term:

$$
\begin{aligned}
H_{sp}^{(f)} = f_1 \sum_n (u_{B,n,x} - u_{A,n,x})(S_{B,n}^y S_{A,n}^z + S_{B,n}^z S_{A,n}^y) \\
-f_1 \sum_n (u_{B,n,y} - u_{A,n,y})(S_{B,n}^x S_{A,n}^z + S_{B,n}^z S_{A,n}^x) \\
+f_2 \sum_n (u_{A,n+1,x} - u_{B,n,x})(S_{A,n+1}^y S_{B,n}^z + S_{A,n+1}^z S_{B,n}^y) \\
-f_2 \sum_n (u_{A,n+1,y} - u_{B,n,y})(S_{A,n+1}^x S_{B,n}^z + S_{A,n+1}^z S_{B,n}^x)
\end{aligned} \tag{15}
$$

where $f_1$ and $f_2$ are, respectively, intra- and intercell magnetoelastic coupling constants. Note that this term is invariant under the following symmetries: $(1, \bar{1}, 2_z, \bar{2}_z, \pm 4_z, \pm\bar{4}_z)$ but not, for instance, under a reflection $\bar{2}_{-xy}$. Such a term $(H_{sp}^{(f)})$ can be written for systems crystallizing, in the paramagnetic phase, in one of the following point groups: $C_{2h}$, $C_{4h}$, $C_{3i}$

or $C_{6h}$ (in Schönflies notation).

We calculate the dispersion relation for $H = T + V_{el} + H_{Heis} + H_{sp}^{(g)} + H_{sp}^{(f)}$ by considering the equations of motion for $\vec{u}_{A/B,n}$, $\vec{p}_{A/B,n}$ and $\vec{S}_{A/B,n}$. The staggered magnetization axis is taken to be parallel to the $\hat{z}$-axis ($\langle S^z_{A,n}\rangle \to \langle S^z_A\rangle$ and $\langle S^z_{B,n}\rangle \to -\langle S^z_A\rangle$). The following characteristic polynomial is obtained

$$
\begin{aligned}
&[m^2\omega^4 - 2m\omega^2(K_1+K_2) + 2K_1K_2(1-\cos k)][\omega^2-\omega_0^2]^2 \\
&-4m\langle S^z_A\rangle^3\alpha[\sin k][\omega^2-\omega_0^2]\omega^3 \\
&-4\langle S^z_A\rangle^6[g_1^2+f_1^2][g_2^2+f_2^2][1-\cos k]^2\omega^2 \\
&-4mJ\langle S^z_A\rangle^4[(B(3+\cos k) - 4\beta(1+\cos k)][\omega^2-\omega_0^2]\omega^2 \\
&+4(\omega^2-\omega_0^2)J\langle S^z_A\rangle^4[3+\cos k][1-\cos k][(g_1^2+f_1^2)K_2 + (g_2^2+f_2^2)K_1][\omega^2-\omega_0^2] \\
&+8J^2\langle S^z_A\rangle^8[g_1^2+f_1^2][g_2^2+f_2^2][1-\cos k]^3 = 0
\end{aligned}
\tag{16}
$$

where $\alpha = g_2f_1 - f_2g_1$, $\beta = g_1g_2 + f_1f_2$, $B = g_1^2+f_1^2+g_2^2+f_2^2$, $\omega_0$ is the dispersion relation of the unperturbed phonon system and $J_1 = J_2 = J$. While the same dispersion relation $\omega = \omega(k)$ is obtained whether we consider right (+) or left (-) circular polarized phonons, Eq. (16) has some important features. It breaks time reversal symmetry ($\Theta$) $\omega(k) \neq \omega(-k)$ due to the terms proportional to $\alpha \sin k$, and also inversion symmetry (I) but respects the product $I\Theta$. The breaking of these two symmetries gives rise to a novel nonreciprocal effect entirely different from the Faraday effect. It translates to the fact that transversal phonon modes propagating in opposite directions have different velocities. Note that this nonreciprocal effect is directly proportional to $\alpha$. If $f_1$ and $f_2$ are zero, there is no such effect.

¿From a closer inspection of Eq. (15), it follows that the three conditions necessary for the above effect to occur in an antiferromagnet are (i) the point group should be one of the following: $C_{2h}$ $C_{4h}$, $C_{3i}$ or $C_{6h}$ [12], (ii) the magnetic ions themselves are not centers of inversion and (iii) the magnetic moment has a component parallel to the main crystallographic axis, $\langle S^z\rangle \neq 0$. The wolframites [13] $CoWO_4$, $NiWO_4$, $FeWO_4$ and $FeNbO_4$ fulfill all

three conditions and are therefore suitable candidates. Their crystal group is $C_{2h}$ and the magnetic unit cell (2a,b,c) is doubled with respect to the chemical unit cell. Another group of candidates are the ilmenites [14] $FeTiO_3$, $NiTiO_3$ and $MnTiO_3$ that belong to the point group $C_{3i}$.

We estimate for $MnTiO_3$, the relative difference

$$\frac{\omega(k) - \omega(-k)}{\omega(k) + \omega(-k)} \tag{17}$$

for the acoustic phonons. We obtain $\omega(k)$ and $\omega(-k)$ by solving Eq. (16) numerically. The values of the constants appearing in Eq. (16) are taken from the literature (see [15] and [16]). The magnetoelastic coupling constants $g_1, g_2, f_1, f_2$ for manganese ions in $MnTiO_3$ are not available in the literature. They can be estimated from first principles by doing an expansion of the exchange term up to the required order [10]. Here, for $g_1$ we assume values similar to those corresponding to other transition metals like Fe. We assume $f_1$ and $f_2$ to be somewhat smaller than $g_1$. This is reasonable given that they define a lower symmetry coupling than $g_1$. $MnTiO_3$ has a spin-wave gap $\Delta(H)$ which varies strongly in an external magnetic field $H$ [15]. We find the novel nonreciprocal effect to be maximal at the resonance frequency of the magnon and the acoustic phonon system which occurs at $\omega \simeq 156$ GHz. At resonance the ratio Eq. (17) is $\sim 10^{-4}$. Note that this nonreciprocal effect occurs already in absence of an external magnetic field. In an external magnetic field the gap $\Delta(H)$ decreases linearly and vanishes at the spin-flop transition $H_c = 5.8$ T. Since the resonance frequency is proportional to the spin-wave gap, a promising experimental set-up in order to measure Eq. (17) would be to sweep the external magnetic field for a fixed phonon frequency. Note that standard measurements of the velocity of acoustic phonons are accurate to $10^{-5} - 10^{-6}$.

*Conclusions* We have studied the possible occurrence of an *acoustic* Faraday effect in antiferromagnets in the absence of an external magnetic field by using a Hamiltonian formulation of the magnetoelastic coupling between phonons and spins and general symmetry arguments. We predict a new boundary- condition induced homogeneous elliptical polarization effect for layered materials like the $ABX_3$-type hexagonal antiferromagnets. The

microscopic analysis of this effect shows that it should be of the same order of magnitude as the acoustic Faraday effect already observed in ferromagnets [3]. We also predict the occurrence of transversal phonon modes propagating in opposite directions having different velocities in certain low-symmetry magnets like the wolframites or the ilmenites. An estimate of the order of magnitude of this effect for $MnTiO_3$ is given. We suggest experiments be undertaken to detect these effects.


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